

Gretchen M. Gallegos Kris A. Surano

Introduction

Because vegetation can be a biological end point for pollutants originally released to the soil, air, or liquids, the sampling and analysis of native vegetation can provide information about the presence and movement of radionuclides in the environment. Vegetation can contribute a radiation dose to humans directly through ingestion or indirectly through human ingestion of the products from animals that have consumed it. DOE guidance states that periodic sampling and analysis of vegetation should be performed to determine if there is measurable long-term buildup of radionuclides in the terrestrial environment (U.S. Department of Energy 1991).

Since 1972, vegetation and foodstuff sampling in the vicinity of LLNL and Site 300 has been part of a continuing LLNL monitoring program designed to measure any changes in environmental levels of radioactivity, to evaluate any increase in radioactivity that might have resulted from LLNL operations, and to calculate potential human doses resulting from direct and indirect ingestion of these products. During 1994, LLNL collected and analyzed samples of native vegetation and wine. In previous years, LLNL collected samples of goat milk and honey but discontinued this because samples became very hard to obtain and the potential doses from those products were very low. By 1993, only one local farm raised goats (and those goats were not kept to produce milk), and only two local honey samples could be acquired. Potential human doses from the remaining foodstuffs—vegetation and wine—are calculated using the monitoring data and dose models presented in Appendix B.

Tritium is the nuclide of major interest in the LLNL vegetation and foodstuff monitoring program because LLNL has historically released tritium to the air both accidentally and in the course of routine operations. Tritium is likely to move into the environment as tritiated water and can be assimilated easily into vegetation and foodstuff. It can contribute to human radiation dose burdens if it is inhaled or ingested directly or indirectly. Although other radionuclides are used at LLNL, our assessments show that only tritium could be present in vegetation in detectable concentrations.

Methods

Our methods for monitoring vegetation and wine are presented in the following sections.



Vegetation

LLNL collects vegetation samples, usually annual grasses, quarterly from fixed locations in the Livermore Valley, San Joaquin Valley, San Ramon Valley, and Site 300, and then analyzes them for tritium. A sampling location designated GARD was added in 1994 at the Livermore site. Location maps are provided in **Figures 10-1** and **10-2**. These locations have been selected so samples would represent vegetation from: (1) locations near LLNL that could be affected by LLNL operations, (2) background locations where vegetation was similar to that growing near LLNL but was unlikely to be affected by LLNL operations, and (3) areas of known or suspected LLNL-induced contamination.

All vegetation sampling is conducted according to written and approved standardized procedures (Tate et al. 1995). Approximately 10% of the sites are sampled in duplicate to comply with quality assurance protocols (Garcia and Failor 1993).

Wine

Wine is the most important agricultural product in the Livermore Valley, representing an approximately \$30-million annual industry. Data since monitoring began have indicated that although tritium concentrations in all wines are low, Livermore Valley wines contain statistically more tritium than do their California counterparts.

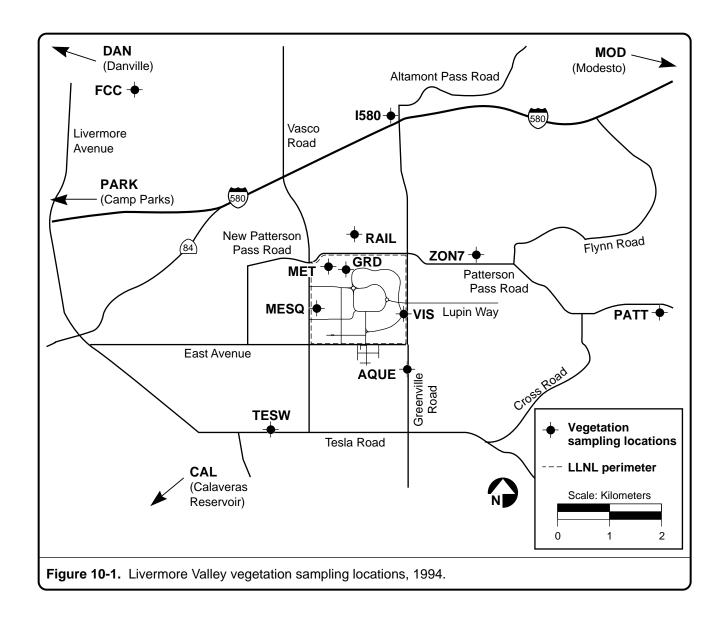
Three types of wine samples of were collected and analyzed for tritium concentrations: wine produced from grapes grown in the Livermore Valley, wines produced from grapes grown in California outside the Livermore Valley, and wines produced from grapes grown in Europe (France, Germany, and Italy). The latter two groups were divided into eight and thirteen wine-producing regions, respectively, and were used as comparative samples.

The wine samples were purchased from local retailers in a variety of vintages and reflect the body of wines locally available to the general public during 1994. The resulting analytical data can be used to estimate the potential tritium dose received by consumers during the year of purchase. The 1994 sampling data cannot, however, be used to indicate how LLNL's operations affected wines produced in 1994. Some time—in some cases, several years—will have elapsed between the harvest of the grapes and the release of the vintage. However, wine sample data can be decay-corrected to its original tritium concentrations (given the number of months that have elapsed between wine production and LLNL analysis) to determine trends and to help determine the impact of LLNL operations during a particular vintage year.

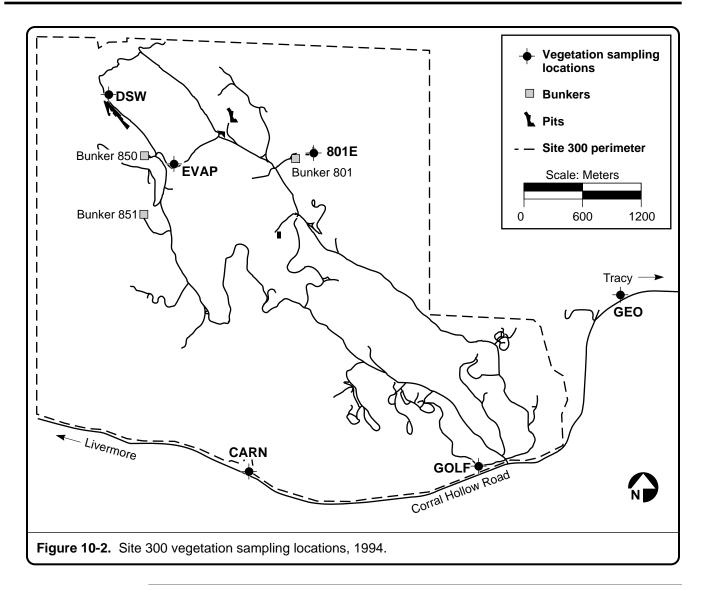
The wine samples were submitted for analysis unopened to avoid airborne tritium contamination. Wines were analyzed for tritium using ³He mass spectrometry in the LLNL Nuclear Chemistry Noble Gas Mass Spectrometry



Laboratory (Surano et al. 1991). We used this highly sensitive method for our wine analysis so that we could determine differences in the tritium content of the samples. Had less sensitive methods been used, such as those employed by commercial analytical laboratories, the tritium content of all samples would be near or below detection limits and no differences would be apparent. Approximately 10% of the total complement of wines were sampled in duplicate to comply with quality assurance protocols.







Results

The results of vegetation and foodstuff monitoring for the Livermore site and Site 300 are presented below.

Livermore

Vegetation

Table 10-1 shows summary tritium data for vegetation collected in the Livermore-site vegetation monitoring program in 1994 (the individual sampling values are presented in Volume 2 of this document). In general, the 1994 tritium levels in vegetation were unchanged from levels measured in 1993 and were lower than levels found in years prior to 1993.



Table 10-1. Tritium in vegetation (in Bq/L), 1994. (a)

Location	Detection Frequency	Median	Interquartile Range	Maximum	Dose (μSv/y) ^(b) Median	Dose (μSv/y) ^(b) Maximum
Livermore site near locations	20/22	8.5	13.0	47	0.041	0.226
Livermore site intermediate locations	12/16	4.0	5.6	19	0.019	0.092
Livermore site far locations	3/20	<1.7	(d)	3.2	<0.008	0.015
Location DSW at Site 300 ^(c)	1/4	<1.9	(d)	340	<0.009	1.636
Location EVAP at Site 300 ^(c,e)	2/2 ^(e)	30	(d)	57	0.14	0.276
All other locations at LLNL Site 300	0/16	<1.8	(d)	<2.1	<0.009	0.010

a See Figures 10-1 and 10-2 for sampling locations.

The vegetation locations were put into three groups for statistical evaluation:

- Near—locations at or within one kilometer of the Livermore-site perimeter.
 Near locations include AQUE, RAIL, GARD, MESQ, MET, and VIS.
- Intermediate—locations in the Livermore Valley removed from the site (1 to 5 kilometers from the Livermore-site perimeter) but close enough and often downwind so that they are still potentially under the influence of tritium releases at the site. The intermediate locations were I580, TESW, ZON7, and PATT.
- Background—locations unlikely to be affected by LLNL operations. Three of
 the background locations (MOD, DAN, and CAL) are more than 25
 kilometers away. The other two (FCC and PARK) are in the Livermore
 Valley but are greater than 5 kilometers from the Livermore site and are
 generally upwind so they are unlikely to be affected by LLNL operations.

The changes in tritium levels between 1993 and 1994 for the vegetation from each of the Near, Intermediate, and Far groups were statistically insignificant.

Because the data for tritium in vegetation were lognormally distributed, the means of the logarithms were compared, using the Tukey-Kramer honestly significant difference (HSD) test. This evaluation showed a significant difference among all three groups, that is, the Near values are significantly different from

Dose calculated based on conservative assumptions that an adult's diet is exclusively vegetables with this tritium concentration, and that meat and milk is derived from livestock fed on grasses with the same concentration of tritium. See Appendix B, Methods of Dose Calculations.

^c Sampling location in known area of contamination.

d Insufficient data to calculate interquartile range.

During the third and fourth quarters, sampling location EVAP was inaccessible due to construction. See Chapter 14, Quality Assurance.



Intermediate, which in turn are significantly different from the Far values. **Figure 10-3** shows the historic averages for the three groups. The highest tritium results for individual vegetation sampling locations were found at AQUE and VIS. These locations are downwind of Sandia National Laboratory, Livermore, and the Livermore site and historically have had higher values than other locations.

Wine

The results from the 1994 wine tritium analyses are shown in **Table 10-2**. Tritium concentrations were within the range of those reported in previous years, and they remained low in wines from all areas.

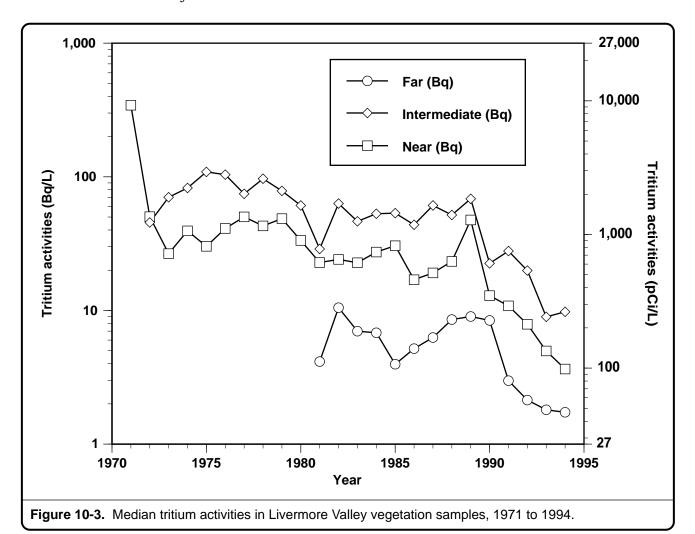




Table 10-2. Tritium (Bq/L) in retail wine, 1994. (a)

	Livermore Valley	California	Europe
Detection frequency	12/12	6/6	4/4
Median	3.60	0.55	1.60
Interquartile range	1.21	0.08	0.38
Mean	4.14	0.57	1.63
Standard deviation	1.81	0.10	0.36
Maximum	8.02	0.68	2.05

Wines from a variety of vintages were purchased and analyzed during 1994. The concentrations shown are not decay-corrected to vintage year.

The data for the 1994 sampling year were analyzed using analysis of variance (ANOVA). The statistical analyses showed that the mean tritium concentration of the Livermore wines sampled was statistically greater than that of both the California (other than Livermore) wines and European wines sampled. The statistical analyses also indicated that there was no significant difference between the means of European and California wines sampled. Multiple comparison tests indicated that the mean levels of the 1994 sampling year data from all areas were not statistically different from those reported for the 1992 and 1993 sampling years. **Figure 10-4**, which shows the results of the wine analyses by sampling year since monitoring began, also shows that 1994 tritium concentrations are among the lowest for all Livermore wines since monitoring began.

Regression analyses and ANOVA of the wine data (when decay-corrected) grouped by vintage year showed tritium concentrations have statistically decreased for all areas since monitoring began, and since 1980. However, the drop in concentrations leveled off for European wines in 1987–1988, in 1990 for Livermore wines, and in 1991 for California wines.

Livermore wines, examined by vintage year, had statistically greater tritium concentrations since 1980 than both European and California wines. This is particularly apparent since 1986 (**Figure 10-5**). However, while vintage wines from Europe exhibited statistically higher tritium concentrations than vintage wines from California from 1980 to 1985, data from more recent vintage years are not statistically different. This indicates that the three distinct data sets discussed in previous annual reports no longer exist; Livermore wines, when decay-corrected and grouped by vintage year, contain higher tritium concentrations than either European or California wines similarly grouped, while European and California wines contain statistically identical concentrations.



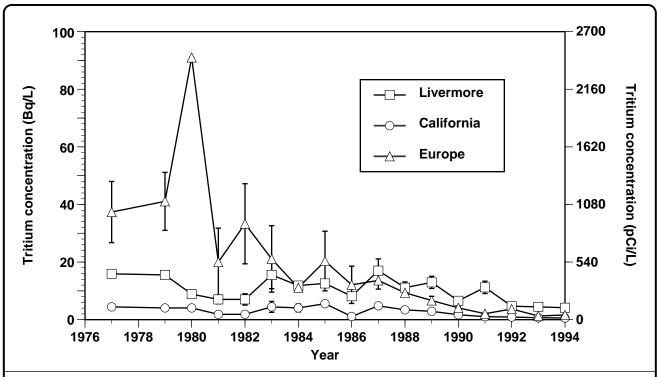


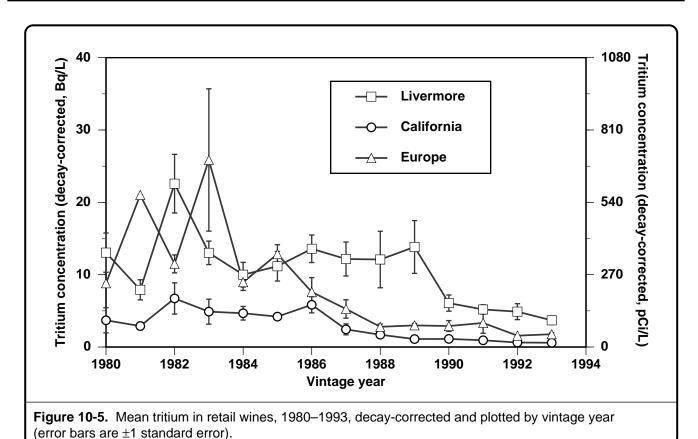
Figure 10-4. Mean tritium in retail wines, 1977–1994, plotted by sampling year (error bars are \pm 1 standard error).

Site 300 Vegetation

Table 10-1 shows summary tritium data for vegetation collected at Site 300 during 1994. Historic values for tritium at Site 300 sampling locations are shown in **Figure 10-6**. Of the six sampling locations at Site 300, four yield results at or near the detection limits. Two locations, EVAP and DSW, yield results above background. Because of construction in the area, the EVAP location could only be sampled twice in 1994. The analytical results for this location were not remarkably higher than for previous years; however, the median is higher because only two samples could be obtained.

As was the case in 1992 and 1993, vegetation samples from location DSW contained the highest tritium values detected. Tritium has been observed in the vegetation of the DSW sampling location since 1971; it is in an area presently being investigated under CERCLA for tritium contamination of ground water. This sampling location is adjacent to a landfill that contains debris contaminated with tritium from past experiments. The landfill area is under continued investigation for tritium in soil and ground water, as described in reports published as part of LLNL's Environmental



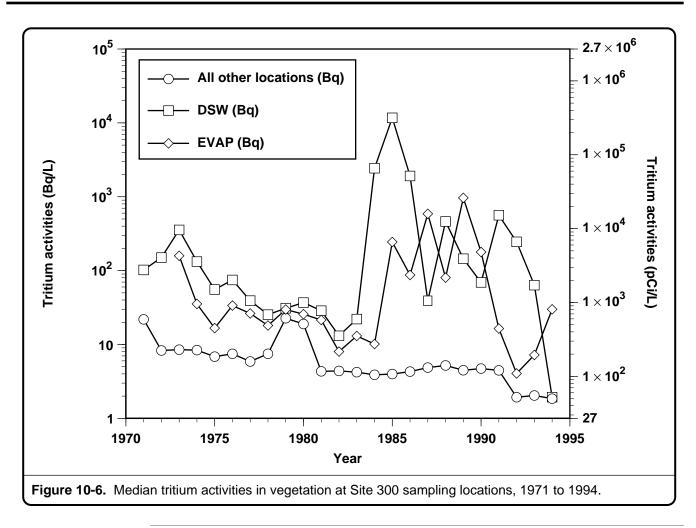


Restoration Program (Lamarre 1989a, 1989b, and 1989c; Taffet et al. 1989a and 1989b; Taffet et al. 1991; Carlsen 1991a and 1991b; and Webster-Scholten 1994). In the past, purge water from samples of ground water monitoring wells was released to the ground at this location. This practice has been discontinued, and LLNL will continue to monitor vegetation in this area to determine whether the change in purge water deposition affects tritium activities in vegetation samples. The location EVAP is near a spring where ground water flows near the surface and evaporates. Some of the ground water near this location arises near the Building 850 firing table where tritium is released to soil (Surano et al. 1995). Consequently, higher than background levels of tritium are measured in vegetation in this area. Evaluation of the 1994 data using the Tukey-Kramer HSD test on the logarithms of the data yielded no significant differences among the locations; however, location DSW and EVAP are significantly different from all other locations when all historic data are evaluated.

Environmental Impact

The environmental impacts of LLNL operations on vegetation and foodstuff monitoring are small and are presented below for the Livermore site and Site 300.





Livermore Site

LLNL impacts on vegetation in the Livermore Valley remained minimal in 1994. The effective dose equivalents shown in **Table 10-1** were derived using the dose conversion factors provided by DOE (U.S. Department of Energy 1988) and thedose pathway model from NRC Regulatory Guide 1.109 (U.S. Nuclear Regulatory Commission 1977). Appendix B provides a detailed discussion of dose calculation methods. The dose from tritium in vegetation is based on the conservative assumptions that an adult's diet consists exclusively of vegetables with the measured tritium concentration, and meat and milk derived from livestock fed on grasses with the same concentration. These assumptions are conservative because most vegetables consumed directly by an adult will not contain tritium at the levels reported (the tritium levels will actually be much less), nor will the livestock actually consume vegetation with the reported levels of tritium. Based on these conservative assumptions, the maximum potential dose (from ingestion of affected vegetation) for 1994 for the Livermore site is $0.23~\mu Sv$ (0.023~m rem).



There are no health standards for radionuclides in wine. However, all the wine tritium levels were far below drinking water standards. In fact, even the highest detected Livermore Valley value (8.0 Bq/L or 220 pCi/L) represents only 1.1% of the California drinking water standard (740 Bq/L or 20,000 pCi/L). Doses from wine consumption can be calculated according to methods for water ingestion, which are detailed in Appendix B.

The corresponding annual dose of the highest detected 1994 Livermore Valley tritium value in wine (8.0 Bq/L or 220 pCi/L) is 0.10 μSv (0.010 μrem), based on the extremely conservative assumption that wine is consumed in the same quantities as water (730 liters per year or 2 liters per day). Using a more realistic wine consumption factor (52 liters per year or 1 liter per week of wine from a single area), and the mean tritium values detected in wines from the three sampling areas, the annual dose from Livermore wine would be 0.0037 μSv (0.00037 mrem), from European wine would be 0.0014 μSv (0.00014 mrem), and from California wine would be 0.0005 μSv (0.00005 mrem). Compared with an annual background dose of approximately 3000 μSv (300 mrem), which includes radon, and a 100- μSv (10-mrem) dose from a typical chest x-ray (Shleien and Terpilak 1984), the potential dose from consuming wine from any area is minute. Therefore, although Livermore wines contained statistically more tritium than wines produced in other areas of California, the effects of the tritium are negligible.

Site 300

In general, LLNL impacts on vegetation at Site 300 for 1994 were insignificant. Tritium levels found in the Site 300 vegetation were comparable to those observed in previous years. With the exception of vegetation from previously identified sites of contamination, the levels were low, near the limits of detection. The areas where tritium is known to be present in the subsurface soil are well delineated and localized.

The calculated maximum potential annual dose from vegetation at DSW, based on the maximum value of 340 Bq/L (9200 pCi/L), is 1.6 μSv (0.16 mrem). This dose, which was not actually received by anyone, is about two orders of magnitude less than a chest x-ray (Shleien and Terpilak 1984). This calculation uses the same conservative pathway modeling assumptions, as described above. In actuality, this dose never would be received because vegetation at Site 300 is not consumed by people or by grazing livestock. In comparison, the calculated potential annual dose from vegetation at all other locations at Site 300 had a median value of <0.009 μSv (<0.0009 mrem; the value is a "less than" value because all measured tritium levels were less than the detection limit). Tritium levels in vegetation at Site 300 will continue to be monitored.